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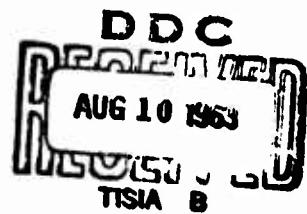
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Technical Report No. 3

Theory of Mechanical Behavior of Heterogeneous Media

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Zvi Hashin



The Towne School of Civil and Mechanical Engineering

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THEORY OF MECHANICAL BEHAVIOR OF HETEROGENEOUS MEDIA

By

Zvi Hashin

The Towne School, University of Pennsylvania, Philadelphia, Pa.

1. Introduction

The purpose of the present survey is to review theoretical methods and results in the field of prediction of bulk mechanical properties of heterogeneous media, in terms of mechanical properties and geometry of constituents. The term heterogeneous medium is here understood as a mixture of discrete homogeneous phases which form regions that are large enough to be regarded as continua. The mechanical behavior of each phase is known in terms of stress-strain relations and at phase interfaces usual continuity conditions on stress vectors and displacements (or velocities) are specified.

The following terminology will be adopted: A heterogeneous medium, consisting of an arbitrary number of phases, will be called a multiphase medium. Its phase geometry is in general random. An important special case of this is a two phase medium. A more special kind of multiphase medium is named suspension and is defined by the restriction that one phase is a matrix in which all other phases are embedded in the form of inclusions. The inclusions may be of arbitrary shapes or specified geometry (e.g. spheres). They may be randomly dispersed in the matrix or form a regular array. Another important special kind of heterogeneous material is a polycrystal which is an aggregate of single crystals, of one kind, whose crystallographic axes

are differently oriented in space, the most important case being that of random orientation. A polycrystal may be regarded as a multiphase material consisting of an infinite number of anisotropic phases. In the case of different kinds of constituting crystals, the term polycrystalline mixture will be used. A phase volume fraction is the ratio of the volume of a phase to the volume of the multiphase body.

Analysis of mechanical behavior of heterogeneous media is important for the following main reasons. First, many materials of technological importance are heterogeneous (e.g. reinforced rubbers and plastics, multiphase alloys, concrete). Second, ability of prediction of properties is prerequisite for design of materials having required properties. Last, but not least, postulates on which continuum theories are based could be critically examined if bulk properties could be predicted on the basis of material structure. The most notable case in point is perhaps the relation between the continuum theory of plasticity and the problem of prediction of plastic behavior of metals in terms of plastic behavior of the constituting crystals.

The starting point of the subject is apparently Einstein's investigation [1]* of the viscosity of a dilute suspension of rigid spheres in a Newtonian viscous fluid, in 1906. Since then a very large number of papers dealing with heterogeneous media of various mechanical properties and geometries has been published. The present survey does not aim at documentation

* Numbers in brackets indicate appended references.

of this field, the contributions to which are scattered in journals of applied mathematics, mechanics, physics, chemistry, metallurgy and polymer science. It is to be regarded as an attempt to review theoretical methods and results from a unified point of view.

There also exists a vast amount of literature on the problem of prediction of dielectric and magnetic behavior, conductivity and diffusivity of heterogeneous media. Since these four problems are mathematically analogous they will be referred to in the following as the dielectric problem. Methods here discussed are mostly applicable to this problem. Work on this problem will be cited only for the purpose of reference to particular methods of investigation.

While the theory of bulk mechanical behavior already incorporates a considerable number of valuable theoretical results, the theory of actual field analysis in heterogeneous media is much less developed and results are available only in very special cases. In fact, achievement in the theory of bulk behavior of heterogeneous media is largely due to the possibility of bypassing the much harder problem of field determination.

2. General Formulation of the Problem

The physical constants of a heterogeneous medium are in general random space functions. Consequently one would expect theories of such media to be of statistical nature. However, most of the work done to date is based on methods of continuum mechanics in which the statistical nature of the

problem has been implicitly incorporated by postulates. Systematic statistical work is of very recent date and only few results have been obtained so far. Statistical formulation of the problem is however indispensable for understanding of the assumptions and limitations of the continuum mechanical approach. While the latter approach has yielded a considerable number of useful results, there are strong indications that in many cases continuation of work will have to be based on statistical methods. In the following both approaches will be discussed.

For complete statistical description of the physical constants as random space functions all their joint probability distributions with respect to any n point system in space, of any configuration, must be known. (For further details here and in the following, see for example, Batchelor [2]). These probability distributions determine the n point averages of the random functions. At present an average is defined in the ensemble sense, by which is meant an averaging process for a fixed system of n points in space over a very large number of specimens of a heterogeneous medium. The theory of bulk behavior is concerned with statistically homogeneous heterogeneous media, by which is meant that all n point averages of the physical constants are independent of the location (but not of the relative configuration) of the n points in space. Any kind of statistical symmetry is defined by invariance of n point averages with respect to space rotations and reflections of the n point system. Mostly one is concerned with statistically isotropic media, in which the

n point averages are independent of any rotation or reflection of the n point system. It is generally postulated that in a statistically homogeneous medium the ensemble average can be replaced by the volume average over a sufficiently large region of one specimen, (ergodic hypothesis). In most investigations statistical homogeneity has been defined by spatial independence of the volume average (see below) and has mostly been described by the terms macroscopic or quasi-homogeneity. The volume region over which the average is taken is necessarily representative of the statistically homogeneous medium and will be referred to as representative volume element (RVE). In the following, unless otherwise stated, the term average will be used for the usual one point, first order volume average, whereas n point volume averages for $n \geq 2$ will be denoted by the usual term correlation functions. The problem of bulk behavior can now be formulated as follows: A statistically homogeneous heterogeneous body of infinite extent is subjected to space constant (ensemble) average strains (or strain rates) $\bar{\epsilon}_{ij} = \epsilon^0_{ij}$. The averages and correlation functions of the physical constants are given. Find the average stresses $\bar{\sigma}_{ij}$ and define the effective stress strain relation by

$$\bar{\sigma}_{ij} = L\bar{\epsilon}_{ij} \quad (1)$$

Conversely the average stresses $\bar{\sigma}_{ij} = \sigma^0_{ij}$ may be prescribed, the average strains sought and the effective stress-strain relation defined by

$$\bar{\epsilon}_{ij} = R\bar{\sigma}_{ij} \quad (2)$$

Here R or L are operators which for heterogeneous media composed of linear elastic phases consist of constant elements and for linearly visco-elastic (including Newtonian viscous fluids) phases of constants multiplied by differential or integral time operators. The material constants entering into the operators will be called effective physical constants. Their number depends on the case treated. For example, for a statistically isotropic elastic medium only an effective bulk modulus and shear modulus are needed. It is in general to be expected that the parameters appearing in the L and R operators are functions of all the averages and correlation functions of the physical constants and it is the purpose of statistical analysis to find these functional relations.

The problem thus presented is very difficult and to date there has been only little work done from this point of view. In one approach the field equations of the random medium (linear differential equations with random coefficients) have been used. Work has apparently been limited to the dielectric case (see for example, Brown [3], Prager [4], Beran and Molyneux [5]. In [3] and [4] series solutions for the effective dielectric constant of two and multiphase media were developed in terms of averages and correlation functions. It was shown that the correlation functions become unimportant only for the case of very small fluctuations of the medium dielectric constant with respect to it's average. In [5] a complete statistical field solution for the electric intensity vector was obtained for the case of small fluctuations). In another statistical approach (Prager [27, 28]) the effective-stress

strain relation is defined by energy (or dissipation) and extremum principles are used to bound the elements of the L and R operators. (The equivalence of the average and energy definition of effective stress-strain relations will be discussed below).

In the statistical approach the correlation functions are considered as given information, in practice they are however very difficult to obtain, especially for higher order. The most easily available information are the one point averages, that is to say, the volume fractions of the phases. It is therefore very useful to consider the following limited aspect of the problem: Given the phase volume fractions and phase stress-strain relations of a statistically homogeneous and isotropic (or other symmetry) heterogeneous medium. What can be found out about the bulk stress-strain relations on the basis of this limited information? Most of the work done and discussed here is of this nature and is based on methods of continuum mechanics. Surprisingly enough a considerable amount of useful information has been gathered within this limited framework. It should however be emphasized that the effective stress-strain relation is in general not determined by such limited information. (except for some very special cases in random geometry or for completely specified phase geometry such as a regular array of identical inclusions) This indeterminacy is a fundamental aspect of the problem. It has, however, been disregarded in many continuum mechanical investigations.

For discussion of the continuum mechanics approach it is useful to cite formulae for average strains and stresses in

terms of boundary values of displacements and tractions. (In the following discussion displacements and strains may be replaced by velocities and strain rates). These formulae are:

$$\bar{\epsilon}_{ij} = \frac{1}{2V} \int_S (u^0_i n_j + u^0_j n_i) dS \quad (3)$$

$$\bar{\sigma}_{ij} = \frac{1}{2V} \int_S (T^0_i x_j + T^0_j x_i) dS \quad (4)$$

Here V is the volume and S the bounding surface of a heterogeneous body, u^0_i and T^0_i are surface values of displacements and tractions respectively, n_i are the components of the outward normal and x_i the cartesian coordinates of surface points. Equ. (3) is based on displacement continuity and small strain definition only, while (4) is based on the stress equilibrium equations, stress vector continuity and zero body forces (it is easily generalized to take account of body forces). In a wide class of homogeneous (space constant properties) media, boundary displacements of the form

$$u^0_i = \epsilon^0_{ij} x_j \quad (5)$$

where ϵ^0_{ij} are space constant strains, or boundary tractions of form

$$T^0_i = \sigma^0_{ij} n_j \quad (6)$$

where σ^0_{ij} are constant stresses, produce space constant strain and stress fields and are thus suitable for experimental determination of stress-strain relations. The theory of bulk mechanical behavior is concerned with heterogeneous bodies of very large extent (compared to phase region size), subjected to such boundary conditions. It follows from (3) that (5)

produces average strains ϵ_{ij}^0 and from (4), that (6) produces average stresses σ_{ij}^0 . Macroscopic homogeneity is defined by postulating that strain and stress averages, for each of conditions (5) and (6), are the same over randomly chosen RVE (see above). This is then equivalent to postulation of space independence of ensemble strain and stress averages. Effective stress strain relations can then again be defined by (1) and (2). If it is desired to proceed only on the basis of the limited information (see above) the definitions (1), (2) are not very useful, because they are now indeterminate. They can be successfully used only in the rare cases where they do become determinate in terms of the limited information and have also been used in approximate treatments, where indeterminacy has been disregarded. Even in determinate cases, such as periodic specified geometry, the use of (1) or (2) is very difficult since the strain or stress fields have first to be found, which in most cases is an impossible task. A more fruitful approach is that of bounding (1) or (2) in terms of the limited information. For this purpose it is very advantageous to define effective stress-strain relations in terms of energy expressions, since then all the powerful energy theorems and in particular variational principles, become available tools. For any heterogeneous elastic body with boundary condition (5), the strain energy is rigorously given by

$$U = \frac{1}{2} \epsilon_{ij}^0 L \epsilon_{ij}^0 V \quad (7)$$

and for boundary condition (6) by

$$U = \frac{1}{2} \sigma_{ij}^0 R \sigma_{ij}^0 V \quad (8)$$

where L and R are given by (1) and (2), respectively. Accordingly the elements of the L or R operators may be bounded by bounding of the strain energy with aid of variational theorems. A number of useful results have been found by this method and in certain cases it could even be shown that the bounds obtained are best possible in terms of the limited information. Energy expressions of type (7) or (8) can also be derived for dissipative heterogeneous media and similar bounding methods then become applicable.

Boundary conditions of type (5) and (6) are, however, too severe for random media. One may superpose on them displacements u'_i or tractions T'_i , respectively, for which (3) or (4) vanish, respectively, without affecting stress or strain averages. If u'_i and T'_i are fluctuating functions whose wavelengths are of the order of phase region typical dimensions and thus very small compared to the whole body dimensions, then macroscopically the heterogeneous body will be insignificantly perturbed. In particular, the additional work done by such u'_i and T'_i will be insignificant in comparison to (7) or (8). All which has been said here also applies to a RVE and thus the equivalence of average and energy definitions of effective stress strain relations is also satisfied in the limit for averages over RVE's of statistically homogeneous media. (For related discussion see Bishop and Hill [62]). Consequently (7) and (8) may be taken as energy densities if V is the volume of a RVE considered as unit volume. Similar considerations apply to dissipative media. In the following results will be

termed exact if the analysis leading to them is based on no approximations beyond the definition of effective stress-strain relations by either averages or energy. While discussion of such results is the main purpose of this survey, a number of useful approximate treatments are also included. In what follows all the heterogeneous media considered are assumed to be statistically homogeneous and unless otherwise stated, also statistically isotropic.

3. Theory of dilute suspensions

A dilute suspension is defined as one in which the fractional volume of inclusions is very small compared to unity. It is generally assumed that in a dilute suspension distances between inclusions are so large that the interactions of their perturbation fields may be neglected. Accordingly the field produced in and around an inclusion when either (5) or (6) is prescribed on the boundary of suspension volume, can be found with sufficient accuracy from the boundary value problem of one inclusion embedded in an infinite matrix where (5) or (6) is prescribed at infinity. Once this problem has been solved the bulk stress-strain relation can be found from (1) or (2) or from (7) or (8) or their analogons. Such inclusion problems can mostly be solved only for spherical or ellipsoidal shape. Accordingly dilute suspension theory is generally limited (with a few exceptions) to such shapes. On the basis of the foregoing assumptions and simplifications, the ensuing stress-strain relations are determinate in terms of volume fractions and phase constants. If the inclusions are

actually of spherical or ellipsoidal, shape the results which will be here discussed are exact for vanishingly small fractional volume of inclusions. Although identical results have been obtained by different methods, the amount of difficulty involved (primarily because of integration over infinite regions) varies from one method to another. In this respect the author believes Eshelby's [6] energy method is the most advantageous since his space integrations are confined to inclusions only. For the sake of compactness discussion will be limited to two phase dilute suspensions. Extension to arbitrary phase number, i.e. inclusions of different kinds, involves no difficulty.

A bulk material constant of a dilute two phase suspension may in general be expressed in the form

$$\frac{M^*}{M} = 1 + ac \quad (9)$$

where

M^* - Bulk material constant

M - Matrix material constant

c - Inclusions volume fraction

a - Non dimensional constant, dependent on matrix and inclusion material constants and inclusion geometry. For spherical inclusions of any sizes, a is independent of geometry.

The range of validity of (9) is usually not more than 1-2%. The primary importance of this expression is in that it gives the slope of the M^* versus c curve at the origin, for a suspension of finite fractional inclusions volume.

When the matrix is a Newtonian viscous fluid, analysis has been carried out only on the basis of the linearized Navier-Stokes equations (neglect of inertia terms). Since viscosity

measurements can be carried out at low Reynolds numbers, this does not seem to be a serious approximation. M^* and M in (9) are now the suspension and fluid matrix viscosity coefficients, respectively. Some of the more important results are the following: Viscous-rigid spheres (in the following the first adjective refers to matrix and the second to inclusions or particles), Einstein [1]. In this case $\alpha = 2.5$ in equ. (9). Viscous-rigid ellipsoids, Jeffery [7]. Viscous-viscous spheres (different viscosity) with surface tension, Taylor [8]. Viscous-viscous spheres (different viscosity) with surface tension, friction and slippage at interfaces, Oldroyd [9]. Viscous-elastic spheres, Fröhlich and Sack [10]. In this case the suspension is viscoelastic and relaxation and retardation times have also been calculated. For a detailed account of theory of viscosity of dilute suspensions, the reader is referred to Sadron [11] and Frisch and Simha [12].

Work on the problem of dilute suspensions in a linearly elastic matrix has begun at a later stage. (For discussion see e.g. Reiner [13]). (For this problem M^* and M in (9) are an elastic modulus of suspension (most convenient-bulk and shear moduli) and matrix, respectively.) The elastic-elastic case (henceforth stands for elastic spherical inclusions dispersed in a matrix, having different elastic moduli) has first been treated by Bruggeman [14]. He found the correct expression for the bulk modulus and an erroneous expression for the shear modulus. The reason of incorrectness has been pointed out by Eshelby [6]. The correct expression for the shear modulus has apparently first been derived by Dewey [15]. (This work seems

to have been overlooked. It was called to the author's attention only very recently). Without knowledge of [15] the more special elastic-spherical voids case was solved by Mackenzie [16] and the elastic-rigid spheres case by Hashin [17]. The results given in [15] were later derived independently by Eshelby [6] and Hashin [18]. Eshelby also gave a method of solution for ellipsoidal inclusions. Results for elastic-viscous spheres suspensions were derived by Oldroyd [19]. This is the counterpart of [10].

The elastic-elastic result occupies a central place in dilute suspension theory. The Einstein formula can be derived from it as a special case, [17], by use of a mathematical analogy between linearized viscous flow and incompressible linear elasticity proposed by Goodier, [20]. On the other hand it may be shown that visco-elastic stress-strain relations of dilute suspensions, consisting of any linearly viscoelastic matrix and inclusions, may be derived by directly applying the correspondence principle (see e.g. [21]) to the elastic-elastic suspension expressions. (Hashin [22]). Special cases of this are the above mentioned results found in [10] and [19].

4. Theory of Finite Suspensions

When the fractional volume of inclusions is finite, suspension theory becomes extremely difficult. It is an interesting fact that the general multiphase medium is more amenable to theoretical treatment than the finite suspension. Obviously results for multiphase media, which will be discussed below, also hold for suspensions. This paragraph, however, is

concerned with treatments which are specific to suspensions and thus in general are not valid for multiphase media.

Work on the theory of viscosity of finite suspensions began very soon after publication of Einstein's result [1] for dilute suspensions and is being continued up to this day. The problem in its full physical generality is very complicated. Such factors as interaction of flow fields around particles, Brownian movement, rotation and collision of particles, non Newtonian and turbulent behavior of the suspension may be regarded as the principal theoretical obstacles. The problem has been mainly considered from its simplest aspect in which theory of bulk viscosity is based on the slow motion steady Newtonian flow field in the presence of many interacting spherical obstacles. Even this idealized problem has proved to be a very formidable one and a satisfactory solution has not yet been found. Discussion or even mention of the overwhelming number of results which have been obtained is beyond the scope of this survey. For such reviews the reader is referred to [12, 23, 24]. ([24] lists close to a hundred different results). Treatments have been generally based on intuitive assumptions which are hard to justify on theoretical grounds. A considerable number of investigations is of a semi-empirical nature, requiring fit of undetermined constants to experimental results. A paper by Kynch [25] will here be mentioned. His treatment is based on potential theory and the flow field in the presence of many interacting spheres of equal sizes is considered in terms of contributions from multipole distributions. Results for suspension viscosity, valid for

moderate fractional volume were obtained. Maude [26] has given an argument whereby the relative viscosity coefficient of a suspension, i.e the ratio of effective suspension viscosity coefficient to fluid viscosity coefficient, should be of the form $1/ (1-2.5c)$, which is the nonlinearized form of the Einstein equation. This expression does indeed agree well with a number of experimental results for moderate concentration. It can however not be valid for the whole concentration range because it takes no account of the statistics of sphere distribution and also because it becomes infinite at $c = 0.4$.

In most of the existing work on suspension viscosity the implicit assumption has been made that the relative viscosity coefficient is a function of the fractional volume of particles only. This assumption cannot be valid for finite suspensions since it disregards the statistical aspect of the problem (compare par. 2). For example: If only fractional volume is specified the fundamentally different cases of concentrated suspension and a porous medium filled with viscous fluid, would fall under the same category. Also experimental results have shown that considerably different values of relative viscosity coefficient can be obtained for the same fractional volume of spherical particles. The importance of the statistical aspect has been emphasized in [25]. It is only very recently that systematic statistical treatment of the problem has begun. Variational bounding techniques with use of correlation functions have been initiated by Prager [27, 28]. Weissberg and Prager [29] treated the spherical suspension problem and gave a lower bound for the effective viscosity coefficient in terms of two point correlation

functions. A closed form lower bound in terms of fractional volume only was obtained, disregarding overlapping of randomly placed spheres. The last result is thus restricted to moderate fractional volume. The major difficulty in statistical treatment is the determination of the correlation functions. It is a characteristic feature of heterogeneous media that the effect of correlation functions increases with the ratio between phase constants, the largest effect being obtained for a rigid phase (or voids) which is the present case.

A lower bound, in terms of fractional volume only, for the effective viscosity coefficient was obtained by Hashin [30] for arbitrary particle shapes by another variational method (see par. 6, below). For moderate fractional volume this bound is only slightly lower than the one derived in [29] for spherical particles.

In contrast with the viscosity problem, the problem of the elastic behavior of a suspension of inclusions in a matrix of different elastic moduli has received little attention. The first investigation known to the author is an approximate treatment by Kerner [31], (see below). From the mathematical point of view the problem is very similar to the simplified viscosity problem, if the inclusions are approximated by spheres. However, this model of an elastic suspension is much closer to physical reality than the corresponding one of a viscous suspension, since all the other complicating factors listed in connection with the latter do not enter. On the basis of the foregoing one has to expect all the theoretical difficulties encountered in the simplified viscosity problem,

if rigid (or empty) inclusions are considered. However, if the ratio's between inclusion and matrix elastic moduli are not too large, restricted treatment is possible. In a paper by Hashin [32], upper and lower bounds for the effective elastic moduli of elastic-elastic suspensions, composed of homogeneous isotropic phases, have been derived by use of the variational principles of minimum potential energy and minimum complementary energy. The following geometric approximation was involved: the suspension was subdivided into composite elements, each of which contained one inclusion. Each composite element was approximated by two concentric spheres, conserving volumes. The method is valid for multiphase suspensions. Closeness of bounds depends on magnitude of ratios between phase elastic moduli, except for the bulk modulus of a two phase suspension where the bounds coincided. At very small fractional volume of inclusions the bounds coincide with expressions for dilute suspensions. The method is rigorously applicable to regular arrays of equal spheres where the above mentioned approximation is not necessary (however, the distance between the bounds is larger than in the approximate treatment). In such cases the particular symmetry of the array determines the symmetry properties of the effective elastic moduli. Krivoglaz and Cherevko [33] treated the elastic spherical two phase suspension with moderate difference between phase moduli, approximately, by a perturbation method.

The important problem of the elastic behavior of fiber reinforced materials can be treated by related variational methods. The first investigation of this kind was undertaken by Hill and

Crossley [34] who considered the case of an elastic matrix reinforced by an equally oriented square array of elastic fibers of equal square cross sections. The reinforced material is anisotropic and has six elastic moduli. Rigorously valid bounds for five of these were obtained. Hashin and Rosen [35] treated the case of equally oriented fibers of hollow or solid circular cross sections, arranged in an hexagonal or a random array. The reinforced material is transversely isotropic and has five elastic moduli. All these moduli were rigorously bounded for the hexagonal case and approximately (by composite element method described above) for the random case, for which coincident bounds were obtained for four of the five elastic moduli.

Finally an approximate method, which is here termed the "smearing out" method, which has been used by several authors, will be mentioned: A typical inclusion is approximated by some regular mathematical shape; at a chosen distance from the inclusion it is assumed that the heterogeneous medium can be replaced by a homogeneous medium whose elastic moduli are the unknowns of the problem. Finite elastic suspensions have been treated in this way by Kerner [31] (also the dielectric case in an adjoining paper) who assumed spherical particles, embedded in concentric spherical matrix shell, then embedded in smeared out suspension material. (In [31] it is implied that the results are independent of matrix shell size. This is not clear to the author). Kerner's results for two phase suspensions have been derived by a different method by Ament [36]. Rosen and Ketler [37] applied similar ideas to

fiber reinforced materials. Budiansky [38] assumed spherical particles embedded directly in smeared out suspensions. (The same method has been applied by Landauer (J. Appl. Phys. 23, 779, (1952)) to the dielectric case. The results compared well with experimental results). Wu [38] treated analogously the case of ellipsoidal particles.

The problem of the linear viscoelastic finite suspension has to the author's knowledge not been theoretically treated. A viscoelastic suspension can be formed in various ways. One such possibility is an elastic matrix containing voids filled with Newtonian fluid, another one is a viscoelastic matrix containing elastic particles. It is evident that there is an enormous number of different kinds of viscoelastic behavior which can thus be obtained. It is a major difficulty of the problem that the form of the visco-elastic stress strain relation is not a priori known. It has been shown(Hashin [22]) that the effective elastic moduli of elastic suspensions and the stress-strain relations of viscoelastic suspensions are related by the correspondence principle. However, this relation holds only for exact results. It is not known whether any information can be extracted from approximate results or bounds for the elastic case.

Finally a curious experimental result will be mentioned. It has been found by Arnstein and Reiner [39,40] that a viscoelastic suspension composed of a cement matrix and sand particles, obeys Einsteins viscosity law for dilute suspensions up to 50-60% fractional volume of sand particles. There is to date no satisfactory explanation of this phenomenon.

5. Theory of Multiphase Media

The general multiphase medium is of completely random geometry. Most of the work in this field has been done for elastic multiphase media, consisting of isotropic phases, and is very recent. A discussion of work for two phase media has been given by Hill [34]. Treatment mostly consists of bound construction for effective elastic moduli by use of the variational principles of the theory of elasticity. Paul [41] derived such bounds on the basis of the variational principles of minimum potential energy and minimum complementary energy. The central problem in bounding methods is to find admissible (compare e.g. Sokolnikoff [42]) displacement and stress fields. In [41], (5) or (6) applied throughout all phase regions, were used as admissible fields. Bound derivation then becomes an elementary problem. It is most convenient to separate the strains and stresses in (5) and (6) into isotropic and deviatoric parts, since then separate bounds for the effective bulk and shear moduli follow immediately. The results are

$$\frac{1}{K^*} = \sum_{r=1}^{r=n} \frac{v_r}{K_r} \quad (10)$$

$$K^* = \sum_{r=1}^{r=n} K_r v_r \quad (11)$$

where K_1^* and K_2^* are lower and upper bounds respectively for the effective bulk modulus K^* , K_r and v_r the bulk modulus and phase volume fraction of the r th phase and n the number of phases. Shear modulus bounds are obtained analogously and the expressions are of the same form as (7) and (8). (In [41] the effective

Young's modulus was bounded instead of the bulk modulus. The Young's modulus bounds are more complicated than the simple expressions (10), (11)). The treatment is rigorous for arbitrary phase geometry. The bounds however are not of much practical value since they become close only for very small differences between the largest and smallest phase moduli. Formulae of type (11) have been used in the older literature as approximate expressions for effective physical constants of multiphase media and have become known as the "law of mixtures".

Hashin and Shtrikman [43,44] constructed improved bounds for arbitrary phase geometry. They derived a class of new variational principles for linear elasticity theory [43,45] which involved the elastic polarization tensor. (The polarization tensor has been implicitly introduced into the theory of elasticity by Eshelby (e.g. [6], [46]). The name has been coined by Kröner [47] in analogy with the electrostatic polarization vector.) A different proof of the variational principles has been given by Hill [48]. The bounds were derived by use of piecewise constant polarization fields. The method involved formal Fourier transform operations whose mathematical rigor has yet to be established. It is however believed that the results are exact in the sense of the statistical formulation of the problem (see par. 2) The results for the n phase medium are too long to quote, and only results for the two phase case will here be written down.

The lower bounds are:

$$K_1^* = K_1 + \frac{(K_2 - K_1)v_2}{1 + \frac{3(K_2 - K_1)}{3K_1 + 4G_1}v_1} \quad (12)$$

$$G_1^* = G_1 + \frac{(G_2 - G_1)v_2}{1 + \frac{6}{5} \frac{(G_2 - G_1)(K_1 + 2G_1)}{G_1(3K_1 + 4G_1)} v_1} \quad (13)$$

Here K , and G denote bulk and shear moduli respectively, 1 and 2 indicate the phases and v stands for volume fraction. The upper bounds are obtained by interchanging 1 and 2 everywhere. (An idea of closeness of bounds may be obtained from the following numerical example: For $G_1 = K_1 = 1$, $G_2 = K_2 = 4$, bounds for bulk and shear moduli of type (10) and (11) are 1.6; 2.5. Bounds of type (12) and (13), for bulk modulus 1.91; 2.21, for shear modulus 1.85; 2.14). The bounds are well confirmed by available experimental results [44]. In the cases of a rigid phase or voids, all the bounds given are of no practical value. In the first case, the upper bound is infinite and in the second the lower bound is zero. It has been shown that the bulk modulus bounds for a two phase medium, given above, are the most restrictive ones that can be obtained in terms of phase elastic moduli and volume fractions. (The dielectric problem has been similarly treated by the same authors, J. Appl. Phys. 33, 3125, (1962). The bounds for the two phase case are also most restrictive). Consequently for improvement of bulk modulus bounds it becomes necessary to take into account second and higher order correlations of the space distribution of the phase moduli. Whether or not the shear modulus bounds are best possible in terms of the information used is at present an open question. The bulk modulus bounds, for two phase materials only, have been rederived by a simpler and mathematically rigorous method by Hill [34]. It is interesting to note that the bulk modulus bounds coincide for

the case of equal phase shear moduli. Hill gave an exact solution for the stress and displacement fields in the phases, for arbitrary phase geometry, in this case. (subject to prescribed average homogeneous isotropic strain or stress). The bounds were derived on the basis of this solution. Hill's solution is the only case of exact field analysis for arbitrary phase geometry and arbitrary relative phase constant magnitudes which is known to the author. The method seems to be restricted to the case which has been solved.

An emulsion of several fluids may be regarded as a multiphase fluid medium. It is obvious that for this emulsion to remain a statistically homogeneous multiphase medium for an appreciable period of time the differences between phase densities must be very small. It is also evident that shear flow will produce orientation effects which will tend to destroy any initially existing statistical isotropy. Another complicating factor is the effect of surface tension at the fluid phase interfaces. It is consequently evident that from the physical point of view the multiphase fluid problem is more difficult than the multiphase elastic problem. For review of various approximate results, see e.g. [23,49]. A method similar to the one used in [44] has been applied by Hashin [30] to bound the effective viscosity coefficient of a fluid emulsion, consisting of Newtonian fluid phases in slow motion. Bounds were obtained for (assumed) initially statistically isotropic and ultimately statistically transversely isotropic (in plane normal to flow direction) phase geometries. Surface tension was not taken into account. The bounds

obtained are of the same order of closeness as bounds of type (12) and (13). In fact the bounds in the statistically isotropic case can be directly obtained from these bounds by letting K_1 and K_2 assume infinite values. The lower bound for a suspension of rigid particles, mentioned in par. 4, follows by assigning infinite viscosity to one phase.

6. Elasticity of Polycrystals

Analysis of bulk elastic behavior of polycrystals is related to the corresponding problem for elastic multiphase media. The constituting crystals are anisotropic homogeneous elastic phases. It is generally assumed that the orientations of the crystallographic axes of the crystals are random or uncorrelated. On the basis of this assumption the polycrystal is taken to be statistically homogeneous and isotropic. The first contribution was apparently by Voigt [50] who assumed that when a displacement of type (5) is prescribed on the boundary of a polycrystal the displacements in all the crystals would be of the same form. Stress vector continuity at crystal interfaces is not satisfied in this approach. The bulk elastic moduli according to this approach are simply expressed in terms of orientation averages of the single crystal anisotropic elastic moduli. The counterpart of this approach is one given by Reuss [51] in which (6) is prescribed on the boundary and the associated stresses are assumed as stress fields in all the crystals. In this approach displacement continuity at crystal interfaces is violated. The bulk elastic moduli so found are expressed in terms of compliance orientation averages of the

single crystals. Hill [52] has shown that the Voigt results are rigorous upper bounds and the Reuss results rigorous lower bounds for the bulk elastic moduli of the polycrystal. It is easily realized that the bounds of type (10), (11) for the multiphase elastic material are of the same kind as the Voigt and Reuss Bounds.

The Voigt and Reuss bounds are close only for slightly anisotropic crystals. Several approximate methods have been devised to obtain better results. A review of some of these has been given by Kröner [53] and Eshelby [54]. Hershey [55] and Kröner [53] have used the "smearing out" method described in par. 4. A single crystal was approximated by an anisotropic elastic sphere imbedded in an infinite homogeneous isotropic elastic medium whose elastic moduli were assumed to be the unknowns to be determined. For cubic crystals the effective shear modulus was thus determined by a third order algebraic equation. (The effective bulk modulus for cubic crystals is rigorously equal to the single crystal bulk modulus). The numerical results obtained are in good agreement with experiments [53].

Hashin and Shtrikman [56] derived improved bounds for the effective elastic moduli of polycrystals of arbitrary crystal geometry by a method similar to the one used by them for elastic multiphase materials. (The same author's treated the problem of the conductivity of polycrystals by a related method, *Phys. Rev.* 130, 129, (1963)). Explicit results were given only for cubic polycrystals. Good agreement with available experimental results was obtained. The following numerical

results are given for comparison: For copper polycrystals, Reuss and Voigt, 4.0; 5.4, Hashin and Shtrikman, 4.48; 4.72 (10^{11} dynes/cm²). The method has recently been extended to polycrystalline mixtures of several kinds of cubic crystals [57]. The bounds thus obtained include as special cases both the bounds of type (12) (13) and the cubic polycrystal bounds.

7. Plasticity of Polycrystals

The fundamental problem in polycrystal plasticity is to predict the macroscopic plastic behavior of a large polycrystalline specimen on the basis of the plastic behavior of the single crystals. Because of the magnitude of plastic strains, crystal lattices are subjected to large rotations when considerable plastic flow sets in. These rotations are usually neglected and it is assumed that the initial macroscopic isotropy of the polycrystal is retained during plastic deformation. The theory is thus restricted to not too large plastic deformation. The problem is conceptually similar to that of polycrystal elasticity, which has been discussed above; it is however much more difficult.

The usually assumed idealized continuum mechanical plastic behavior of single crystals has been postulated by Taylor and his associates on the basis of their experiments [58, 59, 60, 61]. Discussion will here be confined to face centered cubic (f.c.c.) crystals, which is the most widely treated case. These postulates are: (a) The plastic strain in a crystal is due entirely to slip on its twelve slip systems (A slip system is a slip direction in a slip plane. In f.c.c. crystals there

are four slip planes, each of which contains three slip directions. The slip systems form the edges of a regular tetraeder, each edge corresponding to two slip systems in opposite directions) (b) For proportional loading the shear stress operating in a slip system is a function of the shear strain in the same slip system only. The functional relation is the same in all slip systems.

When a polycrystal is loaded from the elastic into the plastic range, single crystals will progressively plastify. It is neither known in which crystals plastic strains appear, nor which slip systems become activated. This may be regarded as the major difficulty of the problem. An account of earlier work has been given by Bishop and Hill [62] and by Cottrell [63]. A well known fundamental investigation is due to Taylor [64, 65] who derived an expression for the uniaxial plastic stress-strain relation on the basis of the following additional simplifications: (c) Plastic strains are large enough to permit neglect of elastic strains (rigid-plastic crystals). (d) The crystals are all in the same state of uniform strain. The problem of the determination of the operative slip systems was resolved by introduction of a minimum principle for the sum of the plastic shear strains in all operative slip systems of a single crystal (on the basis of an assumed principle of minimum plastic dissipation and postulate (b)). After choice of the operative slip systems on the basis of crystal orientation and the minimum principle, an average stress-strain relation of type (1) was calculated. The result is:

$$\sigma = m\epsilon \quad (14)$$

where σ and ϵ are macroscopic uniaxial stress and strain respectively, the functional relationship $\tau(\cdot)$ stands for the shear stress-plastic shear strain relation for any slip system of a single crystal and m is an average orientation factor for which Taylor found the value 3.06. It is evident from (14) that for ideally plastic crystals m is the ratio between macroscopic uniaxial yield stress and single crystal yield shear stress. In an earlier calculation by Sachs [66], who assumed equal uniform stress in all crystals, the value 2.2 (approximately) was obtained for m . In Sachs' calculation displacement continuity at crystal interfaces is violated whereas Taylor's calculation disregards stress vector continuity. Taylor's result is in good agreement with experimental measurements [64, 69].

Taylor's analysis was generalized in a series of papers, retaining the assumption of uniform strain in all crystals. Bishop and Hill [62, 67] (further discussed in [68, 69]) treated the rigid-plastic polycrystal for polyaxial loading and gave results for the yield locus. A principle of maximum plastic work was developed for single crystals and polycrystals, which in their analysis replaced Taylor's postulated minimum principle. In [62] a bounding technique for the yield locus was described, in which the uniform strain assumption was not involved. Expressions for bounds were, however, not given (because the bounds were not sufficiently close, Hill [70]). In [67] the yield locus was calculated on the basis of the assumption that the work done in polycrystal loading can be computed from a uniform strain field. The yield locus obtained is between the Tresca and Mises loci and is in fair

agreement with experimental results for copper and aluminum. The Taylor method was extended to elastoplastic crystals by Lin [71] who described a procedure for calculation of the stress-strain curve in proportional loading on the basis of total (elastic plus plastic) uniform strain in all crystals. The method was extended by Payne [72] to arbitrary loading paths. The elastic anisotropy of single crystals was here neglected. Numerical results for torsion-tension strain history were calculated by Payne and Czyzak [73]. Comparison with experimental results for thin walled cylinders showed considerable discrepancy. In a later application Czyzak, Bow and Payne [74] computed a polycrystalline stress-strain relation for tension-compression strain history.

In another group of papers dealing with polycrystal plasticity, the assumption of uniform strain was not involved and instead various other assumptions were made. Budiansky, Hashin and Sanders [75] considered the problem of initial plastification of polycrystals on the basis of the following approximations: (a) The elastic anisotropy of single crystals is neglected. (b) Each crystal may be regarded as a sphere imbedded in a homogeneous elastic medium ("smearing out" method). (c) It is assumed that local slip occurs in a crystal when the macroscopic resolved shear stress on it's slip system reaches the critical single crystal shear stress (This is strictly true only for isotropic crystals and can be regarded as a good approximation only for slight anisotropy e.g. aluminum). (d) Interaction of the perturbation fields due to the slipped crystals is neglected. Considering only single and

double slip, a stress-strain curve for simple tension was calculated. Because of neglect of interactions the results can be expected to hold only for the very first stage of polycrystal plastification. On the basis of the foregoing it is easy to realize that this work is similar to dilute suspension theory, where here the slipped crystals take the place of embedded inclusions. Kröner [75] devised an approximate method whereby interactions among single crystals could be taken into account, but did not derive explicit results. Kröner's indications for polycrystal stress-strain curve calculation were also based on assumptions (a) and (b) listed above. Budiansky and Wu [77] extended the treatment given in [75]. In their work assumption (d) was not used and slip on all twelve slip systems was taken into account. The "smearing out" method was used by the assumption that in any stage of polycrystal plastification a single crystal was regarded as a sphere embedded in a strain-hardening medium whose characteristics are the unknowns to be determined. Kröner's method for accounting for slipped grain interactions was used. The general method was based on arbitrary loading history and ideally plastic or strain hardening crystals. In the latter case the Prager-Ishlinskii [78, 79] law of kinematic hardening was adapted to single crystal plasticity. The stress-strain curve computed for ideally plastic crystals is below the one derived in [74]: (when taking stress as ordinate and strain as abscissa) For the same uniaxial stress the macroscopic uniaxial strain computed in [77] is $\frac{15(1-\nu)}{7-5\nu}$ times that computed in [74], where ν is the elastic Poisson's ratio of the polycrystal, (Hutchinson [80], reported by Budiansky [81]). For large strains the Taylor limit is obtained.

All the investigations discussed in this paragraph are based on approximations. It should also be noted that none of the theories discussed can take account of the grain size effect of polycrystal plasticity. For discussion and literature concerning this topic, see e.g. [63], p. 124.

8. Conclusion

It has been here attempted to give a unified survey of existing theories of prediction of macroscopic mechanical behavior of heterogeneous media. In spite of the not inconsiderable number of valuable results already available, the whole subject is still in an evolutionary stage. Most of the work discussed is based on the easily available data for a heterogeneous medium, i.e. phase volume fractions and physical constants. Even within this restricted frame there remains much to be done, in particular in the area of more complicated mechanical behavior. For example, to the author's knowledge investigation of elasto-plastic behavior of multiphase materials, except for crude approximations, is not to be found in the literature.

From the general point of view, extension of existing theory to make it possible to take into account correlation functions of heterogeneous media, is obviously needed. It seems that this calls for a systematic statistical theory of the piecewise constant random medium. It also should be noted that the present subject matter deals exclusively with statistically homogeneous stress and strain fields and is only concerned with their averages. The subjects of statistically

nonhomogeneous fields and of microscopic field analysis seem to be virtually unexplored.

Finally, the need for extensive experimental work to check the results already obtained and to provide directions for further investigation should be emphasized.

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